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To cite this article: H S Jeevan and P Gegenwart 2010 *J. Phys.: Conf. Ser.* **200** 012060

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Superconductivity and Magnetism in $\text{Eu}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$

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Abstract. We have studied both single and polycrystalline samples of $\text{Eu}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ with $x = 0, 0.35, 0.45$ and 0.5 by means of electrical resistivity, susceptibility as well as specific heat measurements. Above 40 K, the susceptibility of single crystalline EuFe_2As_2 , measured in fields applied along and perpendicular to the c -axis, is isotropic. In addition to the antiferromagnetic (AFM) spin density wave (SDW) transition at $\simeq 190$ K associated with the FeAs layers, the compound undergoes another magnetic phase transition at 19 K due to AFM ordering of Eu^{2+} moments. For $x = 0.5$, a well defined anomaly in the specific heat related to the superconducting (SC) transition at $T_c = 32$ K confirms bulk superconductivity in this compound. The relative height of the specific-heat jump is larger than the BCS value indicating strong-coupling effects.

1. Introduction

The discovery of superconductivity in Fe-based compounds resulted in a large number of experimental and theoretical studies of the materials containing FeAs layers in a structural unit [1-5]. Similar to the cuprates and heavy fermions, superconductivity in iron arsenides emerges close to the magnetic instability. There are mainly two classes of Fe-arsenides showing superconductivity. RFeAsO crystallizes in the ZrCuSiAs -type structure, composed by alternating (RO) and FeAs layers. The other class of compounds belongs to the well known ThCr_2Si_2 -type structure (so called 122 family), AFe_2As_2 (A: Ba, Sr, Eu or Ca) [6-10] composed of FeAs layers separated by A- layers. Superconductivity in these systems is induced by partial hole or electron doping of the FeAs layers, which otherwise order magnetically. Electron doping has been highly successful for RFeAsO by substitution of oxygen by fluorine or by oxide vacancies, whereas K or Na doping is more successful for AFe_2As_2 compounds.

EuFe_2As_2 belongs to the so-called 122 family. We have recently performed systematic physical property measurements on EuFe_2As_2 single crystals and $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$ polycrystals [9,11]. A very similar magnetic transition related to Fe_2As_2 layers was revealed in EuFe_2As_2 and other AFe_2As_2 systems. EuFe_2As_2 undergoes a SDW magnetic transition at 190 K due to Fe ordering, accompanied by a structural transition [12]. Furthermore it also shows a second magnetic ordering at 19 K due to Eu^{2+} spin ordering. By doping Potassium to the Eu lattice (hole doping), $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$ shows the suppression of the SDW transition and the appearance of a SC phase transition at 32 K. We have also measured the magnetic penetration depth $\lambda(T)$ for polycrystalline samples of $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$, which shows deviations from conventional behavior. A minimum in the penetration depth vs temperature at 4.2 K suggests a strong influence of the Eu magnetic ordering [13] on the superconducting properties. Additionally, our recent Mössbauer spectroscopy measurements clearly establish the coexistence of Eu short-range magnetic order with superconductivity in $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$ samples below 4.5 K [14]. The

application of pressure on EuFe_2As_2 , results in a continuous suppression of the SDW ordering above 2 GPa and a sharp drop in the electrical resistivity, $\rho(T)$, which indicates the onset of superconductivity at $T_c \simeq 29$ K [15]. Interestingly, upon further cooling, the resistivity increases at the Eu ordering temperature, which is nearly pressure independent. Below, we report a detailed study on various concentrations of K doping with particular focus on temperature-dependent specific heat measurements through the SC transition.

Single crystals of EuFe_2As_2 were grown using the Sn-flux method [16]. The polycrystalline K-doped samples were prepared using solid-state reaction; more details on the sample preparation has previously been reported [11]. Electrical resistivity and specific heat were measured using a Physical Properties Measurement System (PPMS, Quantum Design, USA), while for the dc magnetic susceptibility a Quantum Design MPMS system has been utilized.

2. Results and Discussion

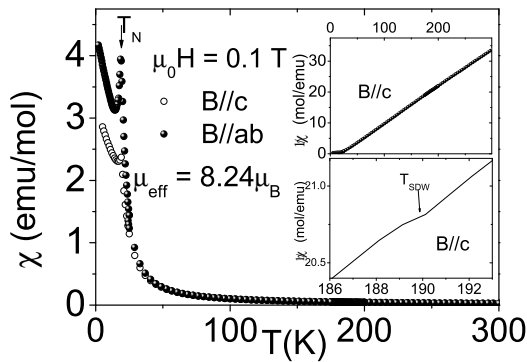


Figure 1. Temperature dependence of the magnetic susceptibility of EuFe_2As_2 single crystals with the magnetic field of 0.1 T perpendicular and parallel to the crystallographic c-axis; drop at 19 K due to AFM of Eu^{2+} moments. Upper and lower insets show Curie-Weiss behavior down to 40 K and small change in slope at 190 K due to SDW ordering, respectively.

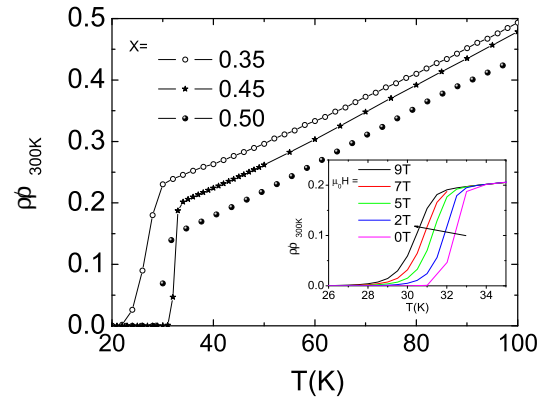


Figure 2. Temperature dependence of the electrical resistivity of polycrystalline samples of $\text{Eu}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, where $x = 0.35, 0.45$ and 0.5 . Inset shows the electrical resistivity for $x = 0.5$ at different magnetic fields up to 9 T. Note the robustness of the SC transition in magnetic field.

Figure 1 shows the temperature dependence of the magnetic susceptibility of EuFe_2As_2 single crystals in two orientations of the applied magnetic field. At high temperatures ($T > 40$ K), there is no difference between χ_{ab} and χ_c , indicating an isotropic susceptibility. In the range of 19 K to 40 K, however, a significant anisotropy in the susceptibility is found (e.g. $\chi_{ab}/\chi_c = 1.6$ at 19 K), suggesting an anisotropic magnetic interaction between Eu spins. At 19 K, a clear drop in χ_{ab} and χ_c indicates the AFM transition. The high-temperature data (above 200 K) follow a Curie-Weiss law (cf. lower inset of figure 1) and can be best fitted with an additional temperature independent susceptibility χ_0 . The so-derived effective magnetic moment for both $H \parallel c$ and $H \perp c$ is close to the theoretical value for Eu^{2+} ($7.94\mu_B$). The Weiss temperature is positive, indicating a predominately ferromagnetic interaction between the Eu^{2+} spins at high temperatures. The Eu^{2+} spins probably align ferromagnetically within the ab planes, but antiferromagnetically along the c-axis, this is recently confirmed by neutron measurements [17]. The lower inset shows the inverse magnetic susceptibility in the temperature range between 185 and 195 K. A small

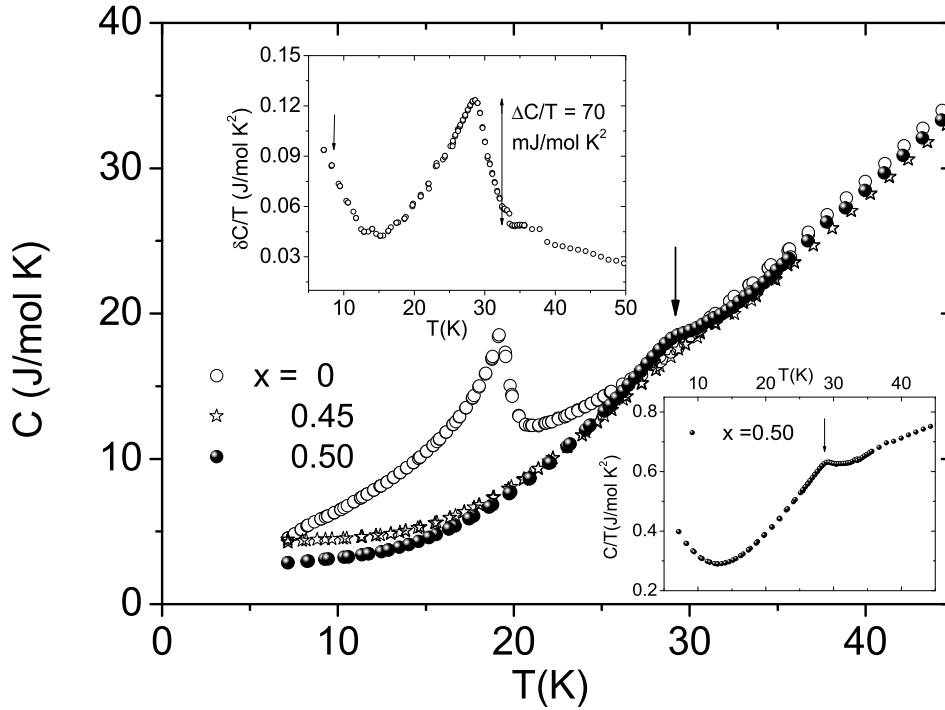


Figure 3. Temperature dependence of the specific heat of polycrystalline samples of $\text{Eu}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, where $x=0.0, 0.45$ and 0.5 . The lower inset displays C_p/T vs T , which clearly shows an anomaly at the SC transition. The upper inset displays the jump height at the SC transition after subtracting a phonon contribution derived from the Sr compound.

change in slope in $1/\chi$ at 190 K could be found for both field orientations (lower inset Fig.1), which is small compared to Ba and Sr 122 compounds due to the strong Eu^{2+} contribution to the susceptibility. This anomaly in $1/\chi$ has been identified to be due to the AFM spin density wave (SDW) transition combined with the structural transition. S. Jiang et al. [18] observed such a SDW transition only after subtraction of the Curie-Weiss contribution of Eu^{2+} moments.

In Figure 2, we present the temperature dependence of the electrical resistivity for $\text{Eu}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ samples with $x = 0.35, 0.45$ and 0.5 respectively. By doping K, we found that the maximum SC transition appears in the sample with nominal composition of $x = 0.45$. In the sample of $x = 0.35$ and 0.5 , the resistivity drop shifts to lower temperatures. The electrical resistivity data exhibit a linear behavior above T_c . The inset shows the magnetic field dependence of the resistivity for $\text{Eu}_{0.55}\text{K}_{0.45}\text{Fe}_2\text{As}_2$. The small shift of the T_c suggests that the SC transition is quite robust against magnetic fields. It has to be noted, that the upper critical fields in these materials are rather high, i.e. of the order of 50 to 60 T [19], in accordance with recently performed high-field resistivity measurements on $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$ [20].

The temperature dependent specific heat for $x=0, 0.45$ and 0.5 is shown in Fig. 3. The broad anomaly at the SC transition ($\simeq 32\text{K}$) arises possibly due to the non-uniform distribution of the dopant concentration. Due to a large phonon contribution at the superconductivity transition a reliable estimation of the normal-state electronic specific heat is difficult. Additionally, there is a magnetic contribution from Eu^{2+} moments, which could not easily be subtracted from the measured data. For this reason we are limited in the experimental determination of $\Delta C_p/T_c$. There is a clear anomaly at the SC transition for $x=0.45$ and 0.5 , which confirms the bulk

nature of the phase transition; the AFM ordering of Eu spins is suppressed after K doping. The SC anomaly is clearer in C_p/T vs T (lower inset of Fig. 3) and a small increase in the electronic specific heat below 10 K is due to the Eu spins. Furthermore, we have subtracted the phonon contribution, derived from the SrFe_2As_2 specific heat (yielding the specific heat increment δC shown in the upper inset of Fig. 3), which gives a rough estimation of the electronic specific heat of $\text{Eu}_{0.55}\text{K}_{0.45}\text{Fe}_2\text{As}_2$. Due to the substantial width of the SC transition, $\Delta C_p/T_c$ and T_c values were determined from this $\delta C/T$ vs T plot. The estimated jump $\Delta C_p/T_c$ is approximately 70 mJ/mol K², resulting in a ratio $\Delta C/(\gamma T_c)$ which is substantially larger than the BCS value of 1.43, hinting at a strong-coupling scenario.

To summarize, the magnetic susceptibility of EuFe_2As_2 single crystals shows two AFM transitions, related to the FeAs and Eu sublattices. The resistivity for various K dopings shows the SC transition with a linear temperature dependence above T_c . The temperature dependent specific heat displays a clear anomaly at the SC transition with a large jump height of almost 70 mJ/mol K².

We acknowledge discussions with C. Geibel, Z. Hossain, D. Kasinathan and H. Rosner. This work was supported by the DFG research unit 960 "Quantum phase transitions".

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